

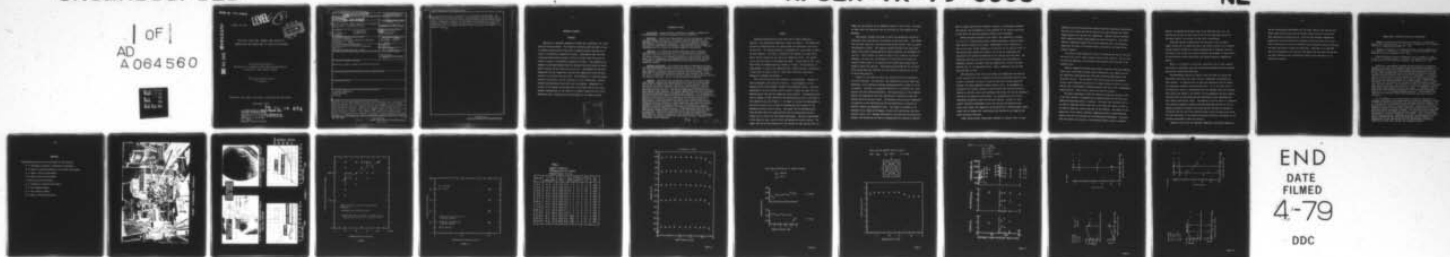
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MULTIPLE IGNITION, NORMAL AND CATALYTIC
COMBUSTION AND QUENCHING OF FUEL/AIR MIXTURES

PRINCETON UNIVERSITY
MECHANICAL & AEROSPACE ENGINEERING DEPARTMENT
PRINCETON, NEW JERSEY 08540

PRINCIPAL INVESTIGATOR
F V BRACCO

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NOVEMBER 1978

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* microns

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and temperature inside and downstream of the catalyst have been made. The dependences of substrate temperature, gas temperature, and gas composition on inlet temperature, reference velocity, and equivalence ratio have been investigated. Homogeneous reactions in the exhaust are observed only at the higher equivalence ratios. Broader interpretation of the variation in exhaust composition with inlet conditions awaits completion of the development of the numerical model.

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ABSTRACT

Ignition of a quiescent propane/air mixture by a stationary, hot, inert particle has been modeled. The tentative conclusion from the model is that an abraded aluminum particle must have a diameter greater than $800\text{ }\mu\text{m}$ to ignite a stoichiometric propane/air mixture. Experimental data show that a burning aluminum particle must be initially larger than $20\text{-}40\text{ }\mu\text{m}$ in diameter to ignite a stoichiometric methane/air mixture. Lean combustion of propane in platinum/alumina/cordierite catalysts has been studied at atmospheric pressure and gas velocities of $5\text{-}30\text{ m/s}$. Measurements of substrate temperature and gas composition, pressure and temperature inside and downstream of the catalyst have been made. The dependences of substrate temperature, gas temperature, and gas composition on inlet temperature, reference velocity, and equivalence ratio have been investigated. Homogeneous reactions in the exhaust are observed only at the higher equivalence ratios. Broader interpretation of the variation in exhaust composition with inlet conditions awaits completion of the development of the numerical model.

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STATEMENT OF WORK

AF FUNCTION - Weapon delivery and defenses, transport, advanced air-breathing engines, aircraft vulnerability and survivability

DEFICIENCY - Insufficient understanding of the basic physical, chemical, and fluid dynamic processes of (1) multiple ignition, propagation, and quenching of flames in spray-air mixtures and (2) of ignition, stability, and efficiency of catalytic combustion. Lack of guidelines for predicting potential flame/detonation quenching techniques and catalytic combustor performance and for solution of existing combustor difficulties.

OBJECTIVE - To clarify the relative importance, and to formulate realistic analytical representation of (1) the mechanisms of multiple ignition by hot metal particles, flame propagation and quenching in fuel-air sprays occurring in air-breathing propulsion system dry bays and fuel tanks, and (2) homogeneous, heterogeneous kinetics and transport processes in catalytic combustion phenomena associated with advanced air-breathing combustion systems.

HOW WORK CONTRIBUTES - Will provide additional understanding and needed realistic analytical modeling of multiple ignition, and flame propagation and attenuation through air-fuel sprays and of homogeneous and heterogeneous high temperature catalytic combustion processes not now available. Will contribute to establishing realistic guidelines and techniques for minimizing ignition probability and maximizing flame quenching and attenuation and for the design of efficient, stable, jet engine catalytic combustors.

APPROACH - Theoretical and experimental studies will be made of basic fluid dynamic, physical and chemical processes of ignition of hydrocarbon fuel sprays by clouds of hot metal particles acting as multiple ignition sources and associated combustion, detonation, and quenching in aircraft fuel tanks and dry bays, and of catalytic combustion associated with air-breathing propulsion systems. The relative importance of gas phase kinetics, heat transfer, mass diffusion, and surface chemical kinetics will be assessed. The practical phenomena will be experimentally simulated. The occurrence or absence of ignition of combustible gas by metal particles will be measured as a function of metal particle size, temperature, and gas-phase composition. Deficiencies in existing mathematical models will be demonstrated and improved models formulated based on experimental data and field observations. Concerning catalytic combustion, various monolithic and packed-bed catalyst candidates for advanced combustor design will be studied over a range of operating conditions characteristic of advanced air-breathing propulsion engines with various hydrocarbon fuels. A two-dimensional model for laminar and turbulent boundary layers with multiple gas and surface reactions will be formulated. Theoretical predictions will be made and compared with measurements of velocity, temperature, and concentrations within the boundary layer above the catalyst obtained by conventional and Raman, absorption and fluorescence laser techniques.

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STATUS

Burning aluminum particles have been used to ignite methane/air mixtures. The experimental apparatus is shown in Figure 1. The combustible gas and an aluminum particle are placed inside the combustion vessel prior to each test. The aluminum particle is suspended on a glass fiber of about 20 μm in diameter. The fiber is attached to the spindle of a depth micrometer, which is mounted on the top of the combustion vessel. The spindle tip is near the center of the combustion vessel. A pulse from the Nd: glass laser heats the aluminum particle causing it to burn. If the particle is large enough, it ignites the combustible gas. Before the test, the particle is moved into the path of the Nd: glass laser beam while observing it through two alignment microscopes.

The events of each test are recorded on two photographs. Examples of these photographs are presented in Figure 2. One photograph is a time exposed picture taken through a window of the combustion vessel. The other photograph is the oscilloscope screen on which is shown the signal from the photomultiplier tube and the combustion vessel pressure. The photomultiplier tube is exposed to light emitted by the burning particle and by the flame in the combustible gas (see Figure 1). In Figure 2, the pair of photographs on the left are from a test in which the aluminum particle burned, but the methane/air mixture did not ignite. The signal from the photomultiplier tube (PM tube) shows the light emission from the burning particle and a bright spot is seen on the time-exposed photograph. The pair of photographs on the right are from a test in which the methane/air mixture ignited. The signal from the PM tube continues to rise because of light emission from the

flame, and the pressure in the combustion vessel is seen to rise. For both of these tests the combustible gas was initially at room temperature and pressure.

Unsuccessful attempts were made to ignite the methane/air mixture by firing the laser when there was no particle on the glass fiber. This showed that the laser radiation, with only the glass fiber present, does not ignite the methane/air mixture. The critical voltage setting on the laser power supply, below which aluminum particles remain unignited, was determined for room air. These voltages are shown in Figure 3 as a function of particle diameter. For each test, the voltage is set only about 50 V above the critical value because it is desired to use no more laser energy than is needed to ignite the particle. This procedure precludes the use of excess laser energy which may interfere with the process of ignition of the gas by the burning particle.

Figure 4 is the format in which the final data from the experiments are to be presented. The data show that methane/air mixtures, under room conditions, can be ignited by burning aluminum particles as small as 40 μm in diameter. Ignition of a propane/air mixture by a stationary, hot, inert particle has been modeled numerically. The concentrations of five chemical species and temperatures have been calculated as a function of time and distance from the particle center. The minimum initial particle temperature for ignition of the combustible gas has been predicted as a function of particle diameter and propane/air ratio. The reader is referred to the reference by Su, Homan and Sirignano, listed in the Publications and Interactions section, for a thorough description of the model and the predictions. Briefly, the conclusions are that an aluminum particle produced by abrasion

must be larger than 800 μm in diameter to ignite a stoichiometric propane/air mixture; and the propane/air ratio ignitable by the smallest particles is that ratio for which the propane oxidation rate is maximum.

During the period covered by this report, lean combustion of propane in a 25 \times 25 \times 76 mm platinum coated, alumina washcoated, cordierite honeycomb substrate catalyst was studied. Measurements of the uniformity of fuel to air ratio, of gas temperature and velocity at the catalyst inlet, of the pressure drop through the catalyst, and of the gas temperature and composition both inside and downstream of the catalyst have been made. The operating conditions over which these measurements have been made are 1 atmosphere pressure, 500-800°K inlet gas temperature, 5-30 m/s inlet gas velocity and .19-.32 equivalence ratio. Table 1 summarizes the runs that have been made to date.

The uniformity of the fuel to air ratio, gas temperature and velocity at the catalyst inlet over a substantial part of the honeycomb catalyst cross section is necessary in order to avoid hot spots which could destroy the catalyst and radial gradients between catalyst cells. An arrangement of stainless steel screens, upstream of the catalyst, has been devised which results in $\pm 1\%$ fuel to air ratio uniformity and within $\pm 5\%$ velocity uniformity over 80% of the catalyst cross section. By insulating the test section, temperature uniformity within $\pm 1\%$ has been achieved. Figures 5-7 show typical uniformity measurements, where the velocity was measured with a pilot static probe, the temperature was measured with a chromel-alumel thermocouple and the propane fuel concentration was measured by gas sampling and flame ionization detection.

Eight chromel-alumel thermocouples imbedded in catalyst cells at eight

different axial positions provide catalyst substrate temperature measurements. The cells are blocked off from the fuel-air flow and therefore the thermocouples measure the catalyst wall temperature. Typical catalyst substrate axial temperature profile measurements are shown in Figure 8. The radial substrate temperature profile has also been measured for a few operating conditions and found to be uniform within a few percent over half of the catalyst diameter.

The pressure drop through the catalyst is measured not only at the inlet and outlet but also at three locations inside of the catalyst. This is done by inserting hypodermic size stainless steel pressure taps into one of the catalyst cells.

Using an expansion quenched, water cooled, stainless steel gas sampling probe with a side mounted chromel-alumel thermocouple, gas composition and gas temperature measurements have been made inside and downstream of the honeycomb catalyst. The gas samples have been analyzed for hydrocarbons using a total hydrocarbon analyzer with a flame ionization detector, for CO and CO₂ with a nondispersive infrared analyzer, and for O₂ with a paramagnetic oxygen analyzer. Some of these results are shown in Figure 8.

The probe measurements inside the catalyst are made by drilling a hole, in the same catalyst, of progressively lengthened depth into which the gas sampling-thermocouple probe is inserted. The probe cross-sectional area is an order of magnitude smaller than that of the probe hole which in turn is an order of magnitude smaller than that of the catalyst. The effect of the probe hole on the catalyst substrate temperature profile is monitored and found to be within the accuracy of the thermocouple measurement. The probe hole does affect the velocity in the shortened catalyst cells of interest;

however, by matching the pressure drop in the shortened cells (i.e. by lowering the overall air flow rate) to that in the equivalent section of the whole catalyst, the velocity in the cells is maintained.

Using this method to probe inside the catalyst is not the same as simply cutting off the downstream end of the catalyst because of the effects of heat transfer through the catalyst substrate and of radiation losses at the catalyst exit, where the respective absence and presence of these effects in the shortened catalyst would alter the catalyst substrate temperature profile.

While it is difficult to draw final conclusions until a more complete set of data is available, some very interesting phenomena have been observed and they are briefly described below.

The measurements plotted in Figure 8 show the effect of varying the equivalence ratio while the inlet velocity, temperature and pressure are kept constant. As expected both the substrate temperature and the exhaust gas temperature increase with equivalence ratio. It is also seen that as the equivalence ratio is increased more fuel is consumed inside the catalyst. More interesting though is the behavior of the homogeneous gas phase exhaust reactions which go from being non-self-sustaining to self sustaining over this range of equivalence ratios. The behavior of the CO curves is consistent with accepted homogeneous propane oxidation mechanisms whereby the rate of propane consumption by means of OH attack and the resultant CO production is greater than the rate of CO oxidation which is also by means of OH attack. Note the measurement at the bottom of the probe hole which corresponds to the gas phase measurement inside the catalyst.

Figures 9 and 10 show the substrate temperature, gas phase temperature

and the concentration measurements for the whole catalyst case and the case with a probe hole half way into the catalyst where the inlet velocity has been reproduced by matching the Δp . Note that the substrate temperature profile over the first half of the catalyst is the same within 5% for both cases indicating that the inlet conditions and surrounding catalyst conditions have been kept nearly constant for both cases. From this it is concluded that the measurement at the bottom of this probe hole does equal the average gas phase temperature and concentration values at the same point in the undisturbed catalyst.

PUBLICATIONS, PRESENTED PAPERS AND INTERACTIONS

"Numerical Predictions of Conditions for Ignition of a Combustible Gas by a Hot, Inert Particle", by Y. P. Su, H. S. Homan and W. A. Sirignano, submitted to Combustion and Flame, Nov. 1978.

"Progress of Reaction in a Honeycomb Catalyst: CO/Air Combustion", by P. M. Walsh, D. A. Santavicca, B. Kim and F. V. Bracco, presented at EPA Workshop on Catalytic Combustion, Oct. 1978.

"High Temperature Catalytic Combustion", by P. M. Walsh, C. Bruno, B. Kim, D. A. Santavicca and F. V. Bracco, to be presented at the Eastern States Section of the Combustion Institute, Fall Meeting, Nov. 1978.

Scientists in government, industry and universities have expressed interest in our work. The Allegheny Ballistics Laboratory, Hercules, Inc., of Cumberland, Maryland and Professor George Brown of the University of Rhode Island have requested our publications and information on our work. In June, 1978, Professor Wojciki of the University of Warsaw in Poland visited Princeton University and was shown our experiments. In November, 1978, Dr. Edmond Kyser of Systems Industries visited our labs to show us how the ink jet printing technique may be used to create monodispersed fuel sprays for future experiments in our AFOSR program. After the paper listed above was presented at the Combustion Institute Meeting in April, 1978, Professor Dabora of the University of Connecticut requested information about how we ignite metal particles with a laser; his experiments involve laser-induced gas breakdown to ignite fuel sprays. In February, 1978, Dr. Yurii Frolov and Professor George Manelis of the Institute of Chemical Physics, U. S. S. R., visited Princeton University. They brought to our attention some research on metal particle burning described in Russian literature.

Dr. W. Pfefferle, one of the pioneers in catalytic combustion, is extremely interested in our program and has visited our laboratory a number of times for detailed discussion of our catalytic combustion program. Dr. R. M. Kendall of the Aerotherm Division of Accurex has also visited us for discussion of our catalytic combustion program. Interest has also been shown by Cerkanowicz of Exxon Labs, Robben of Lawrence Berkeley Labs, Hanak of Matthey Bishop and Sowards of Dupont. The Fall Meeting of the Western States Section of the Combustion Institute, October 1977, half of which was devoted to catalytic combustion, was attended. Visits to other laboratories studying catalytic combustion were made: Aerotherm Division of Accurex (Kesselring) and Lawrence Berkeley Laboratories (Robben). A presentation of this work was given at the EPA Workshop on Catalytic Combustion, October 1978, and was very favorably received.

PERSONNEL

The following personnel have contributed to this research:

- W. A. Sirignano, Professor, Co-Principal Investigator
- F. V. Bracco, Associate Professor, Co-Principal Investigator
- H. S. Homan, Research Staff Member
- P. M. Walsh, Research Staff Member
- C. Bruno, Research Staff Member
- D. A. Santavicca, Research Staff Member
- Y. P. Su, Graduate Student
- B. S. Kim, Graduate Student
- S. O. Morris, Technical Specialist

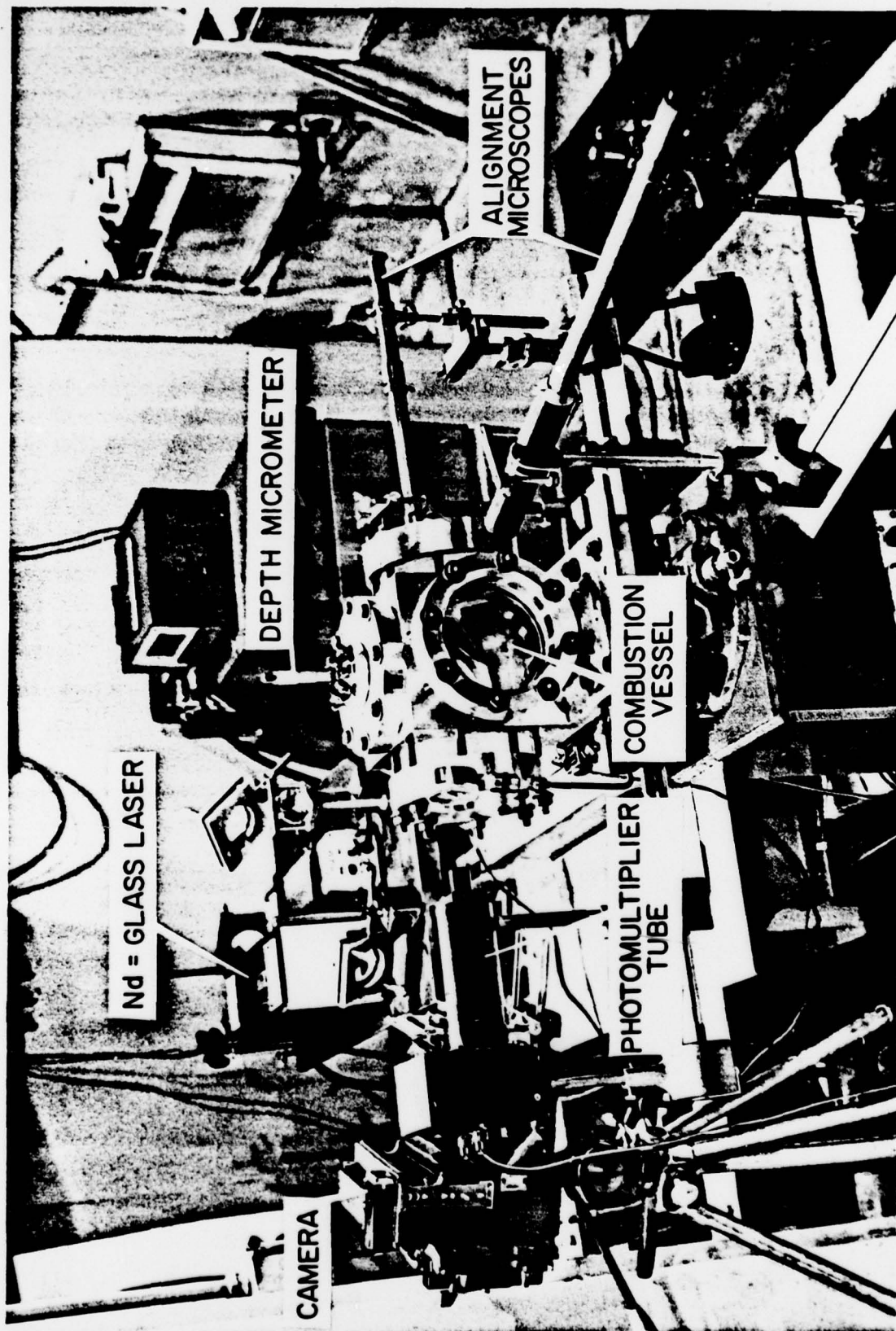
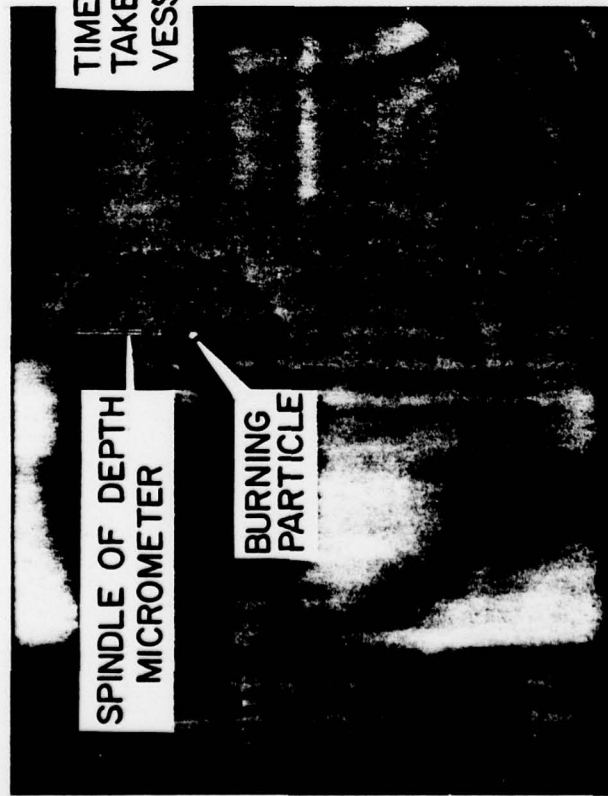


FIGURE 1 EXPERIMENTAL APPARATUS



$\phi = 0.7$, PARTICLE DIAMETER = $45 \mu m$
GAS NOT IGNITED



$\phi = 1.0$, PARTICLE DIAMETER = $40 \mu m$
GAS IGNITED

TIME EXPOSURES
TAKEN THROUGH
VESSEL WINDOW

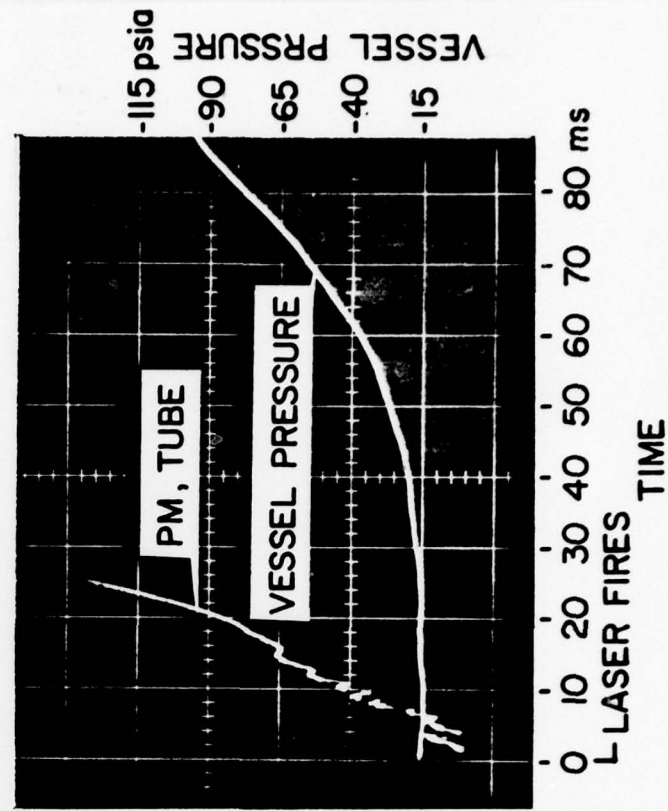
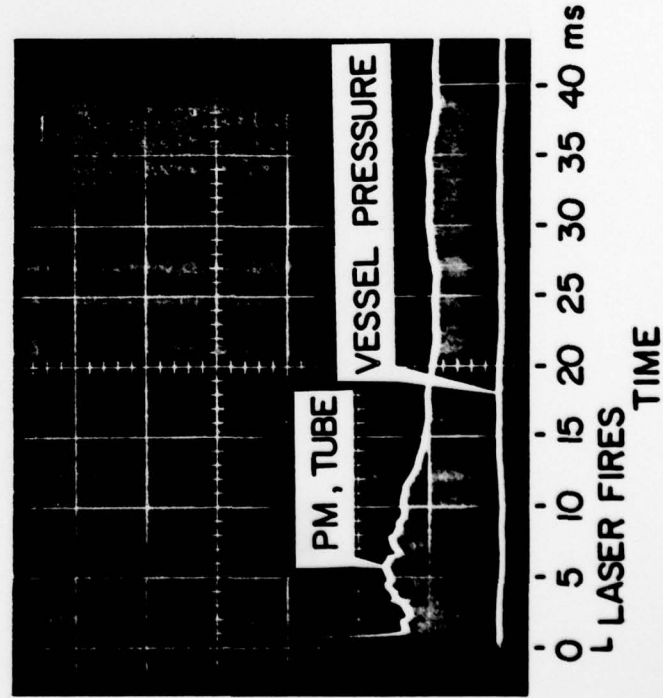
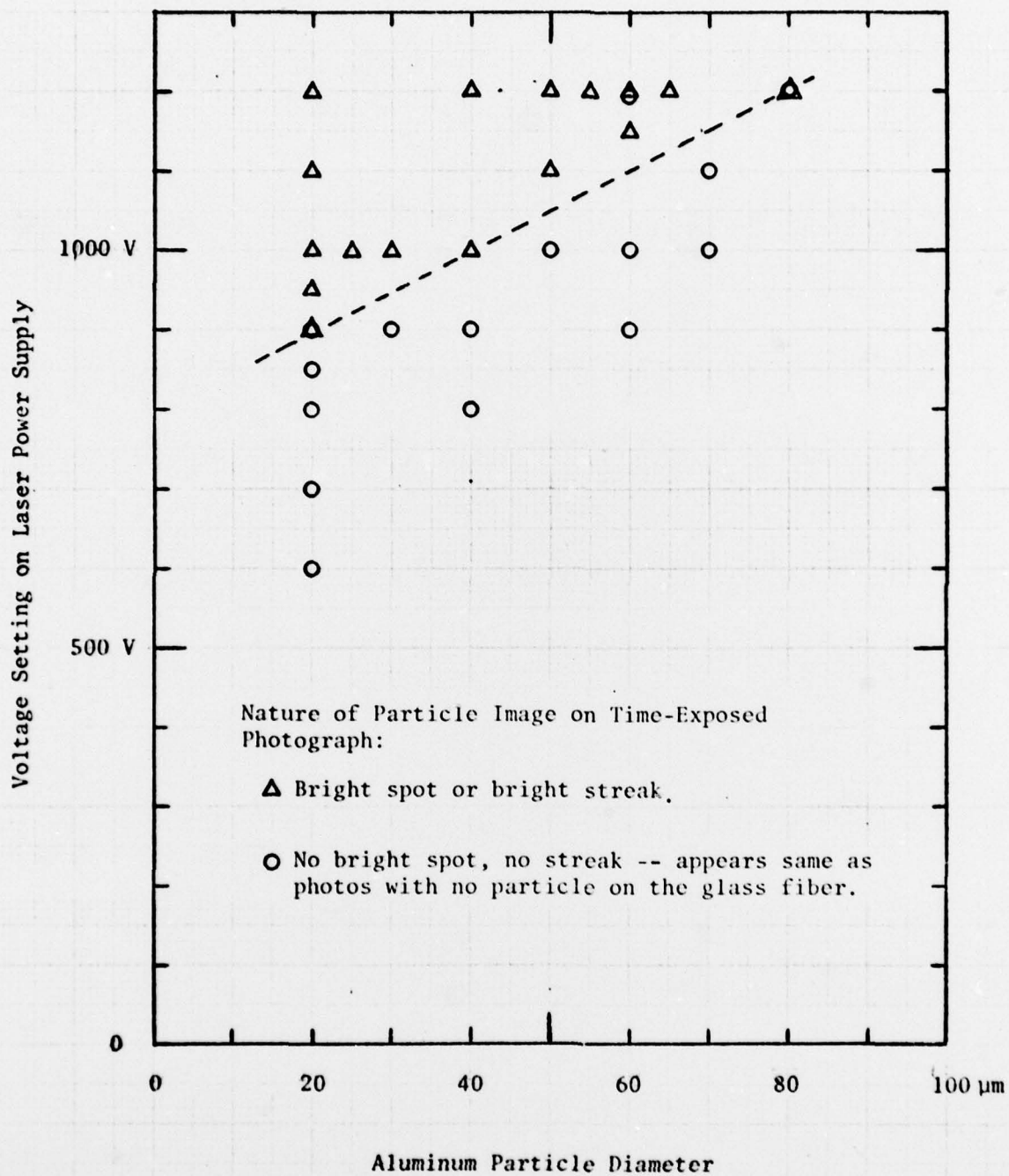


FIGURE 2



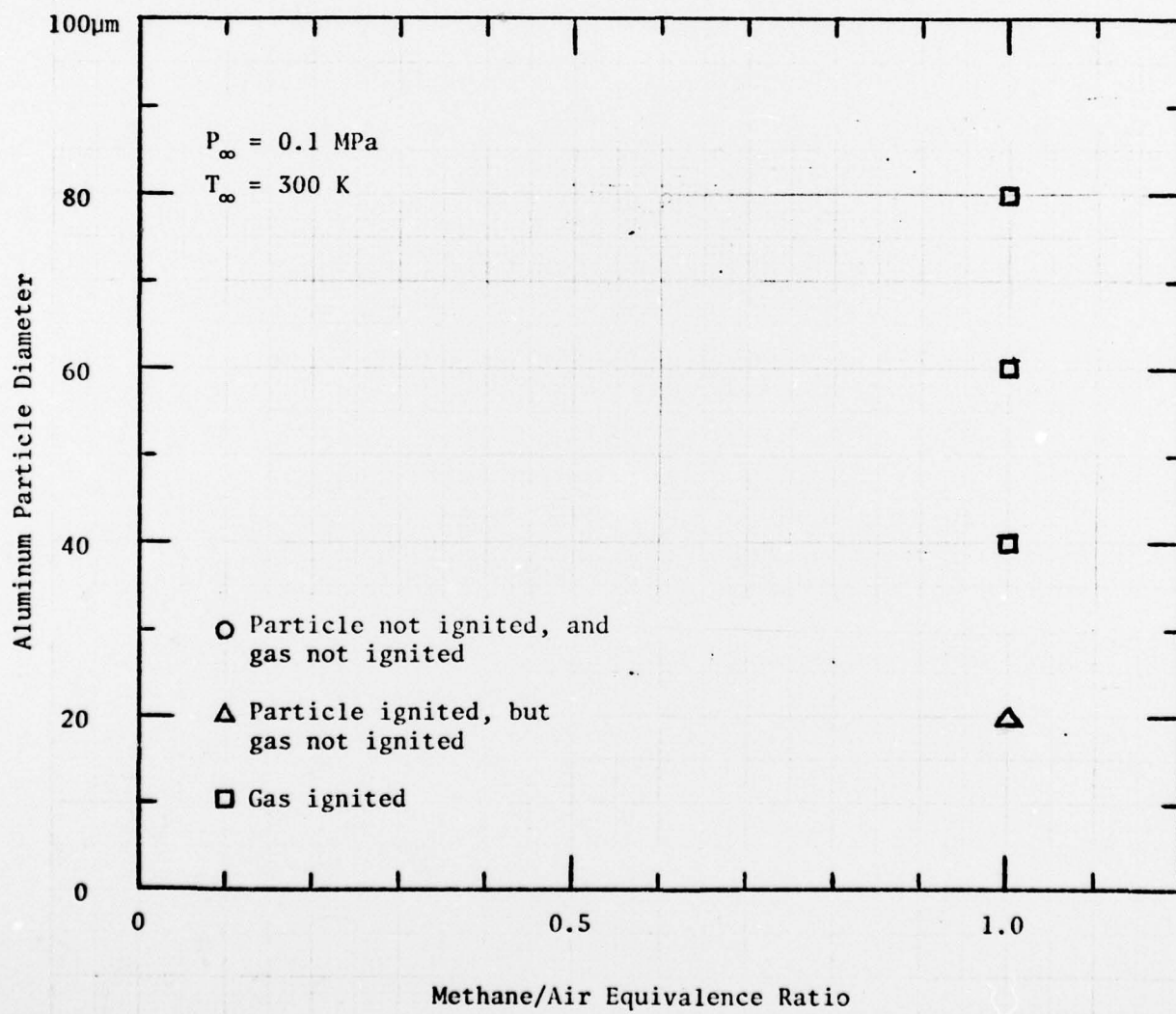


FIGURE 4

TABLE 1

C_3H_8 /Air Combustion
Pt/ Al_2O_3 /Cordierite Catalyst
Overall Length = 76.3 mm

| Run No. | l (mm) | P_{in} (kPa) | T_{in} (K) | ϕ | u_{ref} (m/s) | $(H_2O)_{in}$ (mol%) | $\Delta P/P_{in}$ (%) | T_{ad} (K) |
|---------|-------------|-------------------|-----------------|--------|--------------------|-------------------------|--------------------------|-----------------|
| *11-2 | 76 | 110 | 750 | .30 | 6 | 1.3 | 1.2 | 1480 |
| 11-3 | 76 | 110 | 750 | .29 | 12 | 1.2 | 2.5 | 1460 |
| 11-4 | 76 | 110 | 750 | .32 | 24 | 1.4 | 5.6 | 1520 |
| 11-5 | 76 | 110 | 750 | .19 | 9 | 1.5 | 1.3 | 1230 |
| 11-6 | 76 | 110 | 750 | .22 | 9 | 1.6 | 1.4 | 1300 |
| 11-7 | 76 | 110 | 750 | .25 | 9 | 1.7 | 1.6 | 1370 |
| †11-8 | 76 | 110 | 750 | .28 | 9 | 1.6 | 1.8 | 1430 |
| 11-9 | 76 | 110 | 650 | .28 | 9 | 1.2 | 1.9 | 1350 |
| 11-10 | 76 | 110 | 700 | .28 | 9 | 1.2 | 1.9 | 1390 |
| ‡11-11 | 76 | 110 | 800 | .27 | 9 | 1.3 | 1.8 | 1450 |
| 11-12 | 76 | 200 | 750 | .29 | 12 | 1.2 | 1.3 | 1460 |
| *11-13 | 38 | 110 | 750 | .30 | 4(6) | 1.2 | 0.7 | 1480 |
| †11-14 | 38 | 110 | 750 | .29 | 5(9) | 1.1 | 0.9 | 1460 |
| ‡11-15 | 38 | 110 | 800 | .28 | 6(9) | 1.4 | 0.9 | 1480 |

$u = 8 \text{ m/s}, \phi = 0.28$

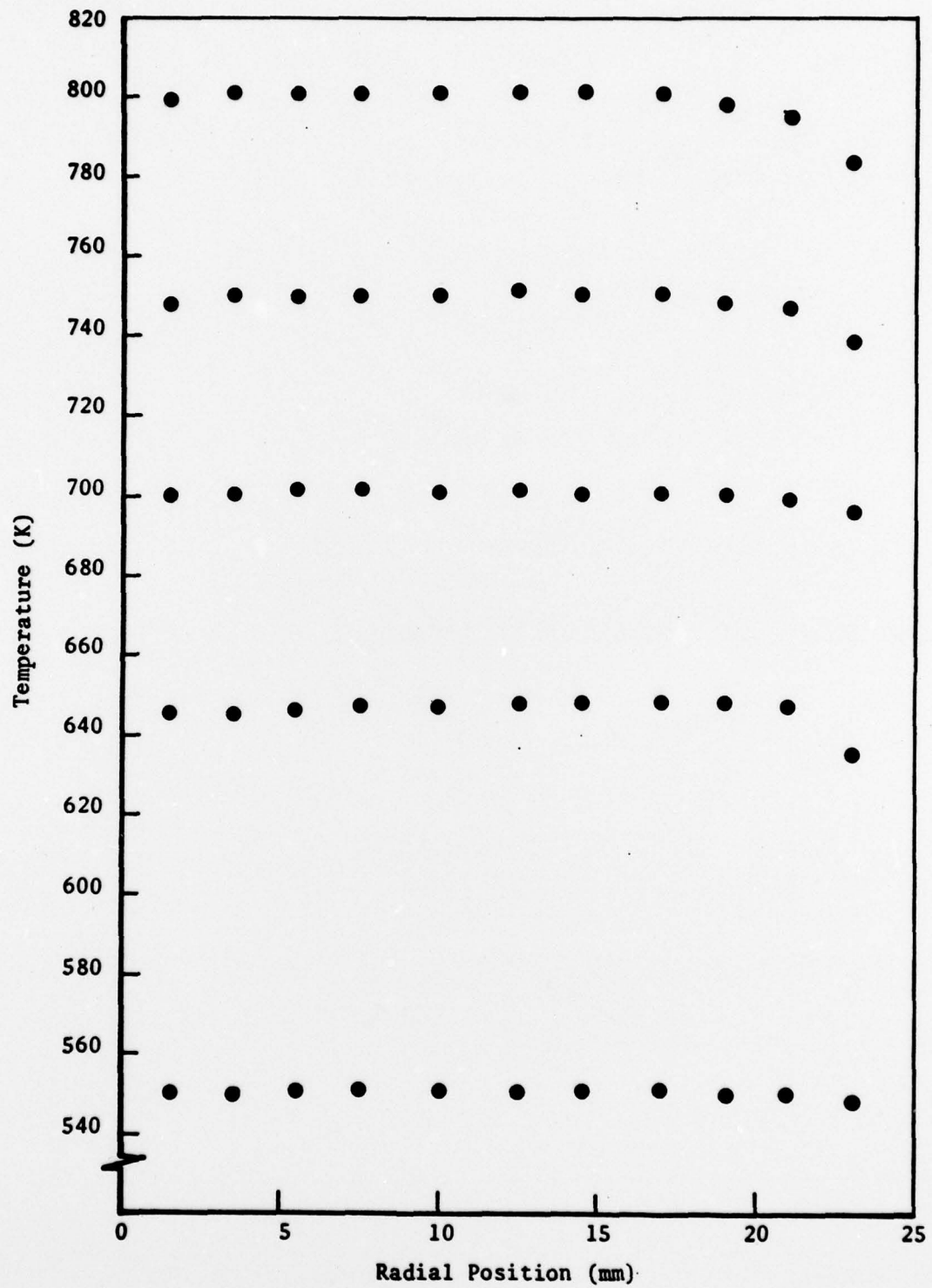


Figure 5

Inlet C_3H_8 Concentration vs. Radial Position

$P_{in} = 100 \text{ kPa}$

$T_{in} = 750 \text{ K}$

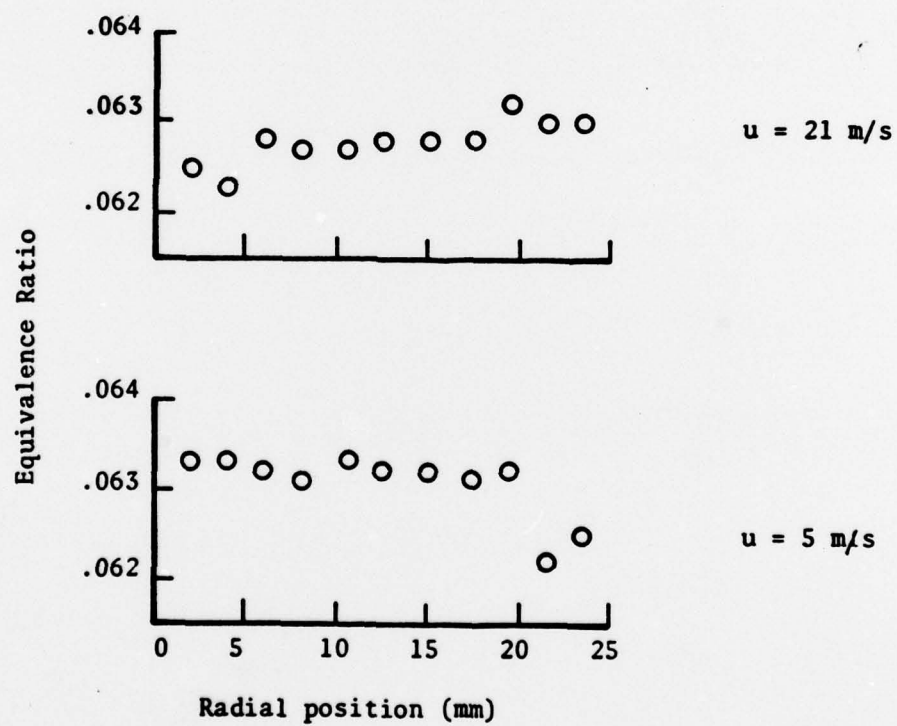


Figure 6

Inlet velocity profile with new screen.

Fuel: C_3H_8 $T_{in} \approx 750^\circ K$ $\dot{V} = 3 \text{ scfm}$

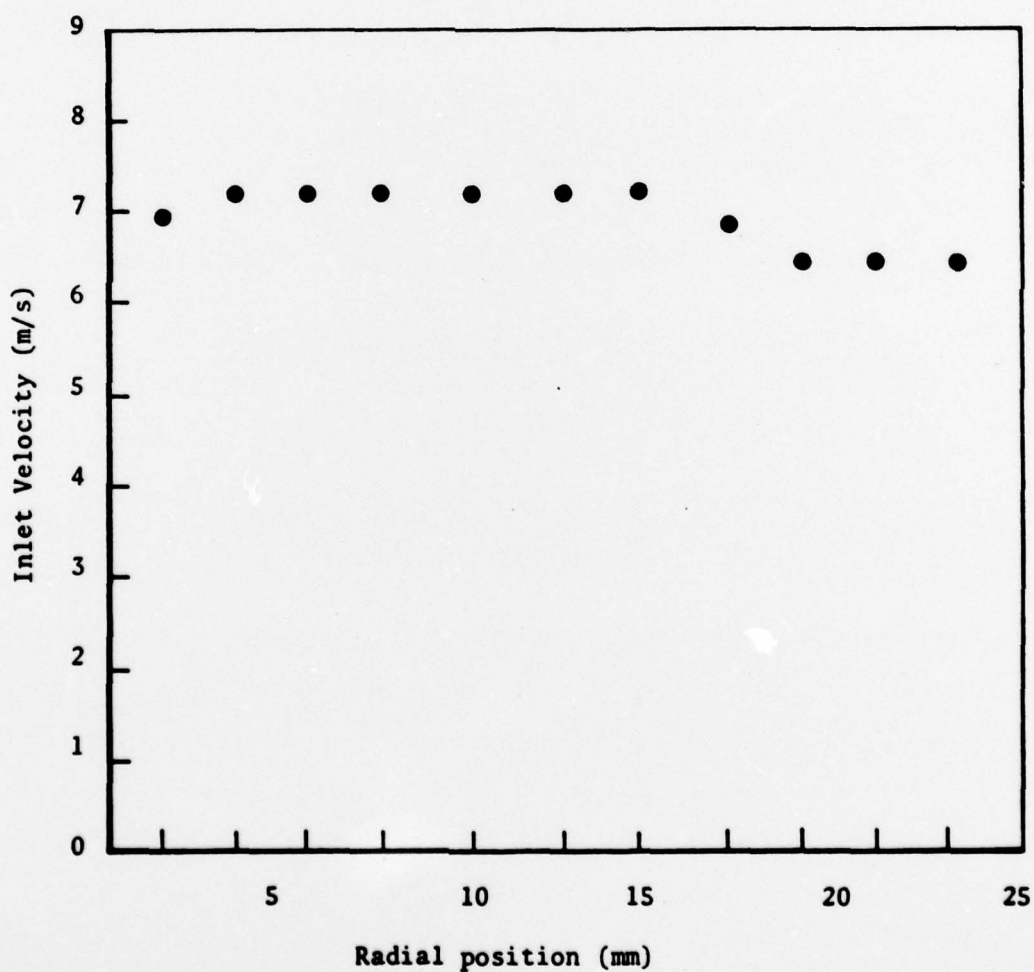
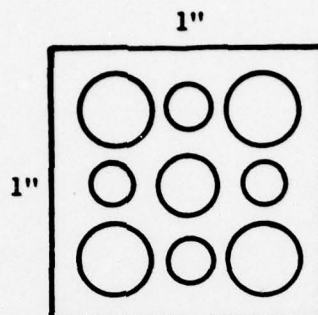


Figure 7

Runs 11 - 5, 6, 7, 8 - C_3H_8

$P_{in} = 110 \text{ k Pa}$

$T_{in} = 750 \text{ K}$

$H_2O = 1.6 \pm 0.2 \text{ mol } \%$

$u_{ref} = 9 \text{ m/s}$

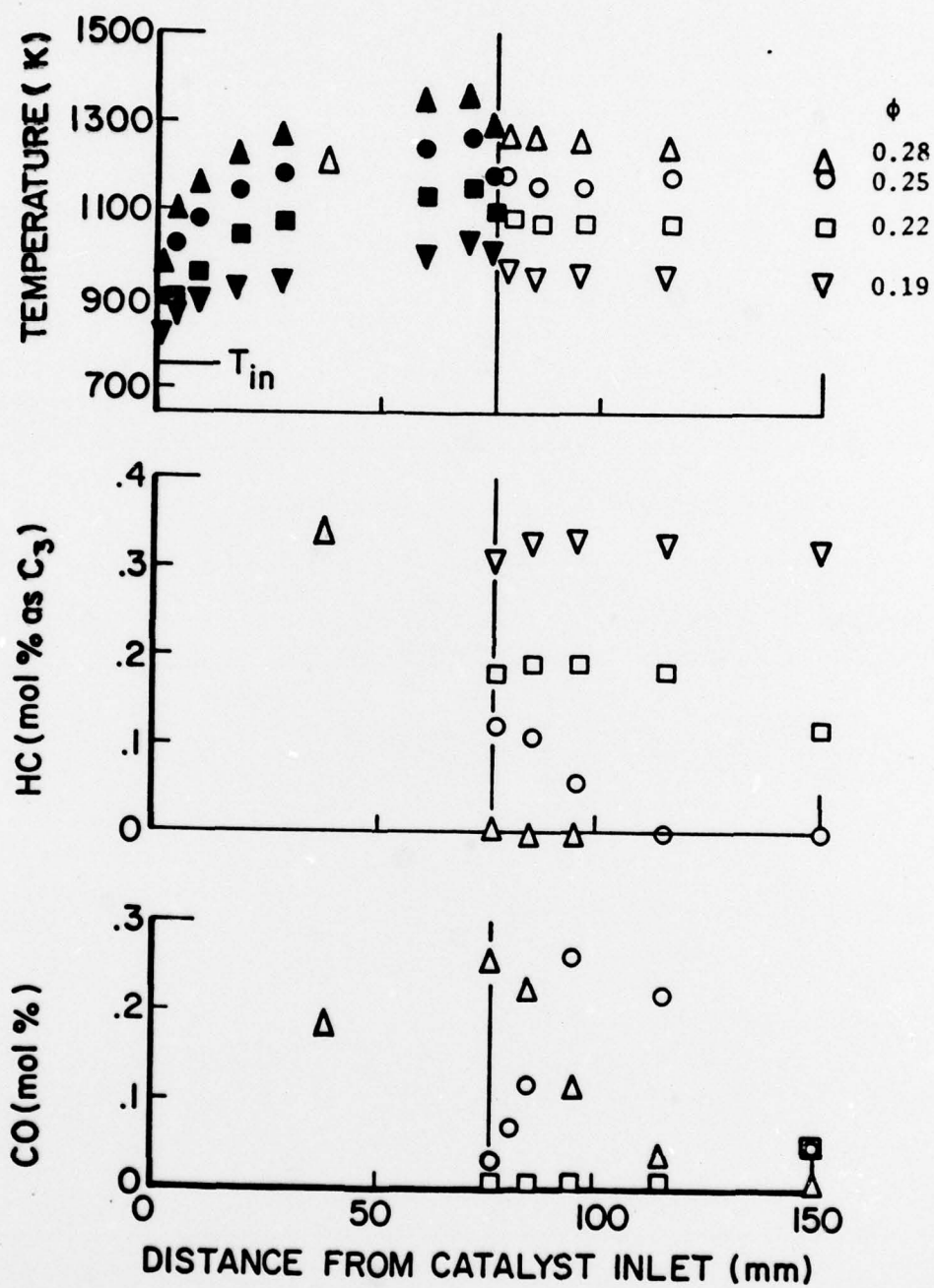
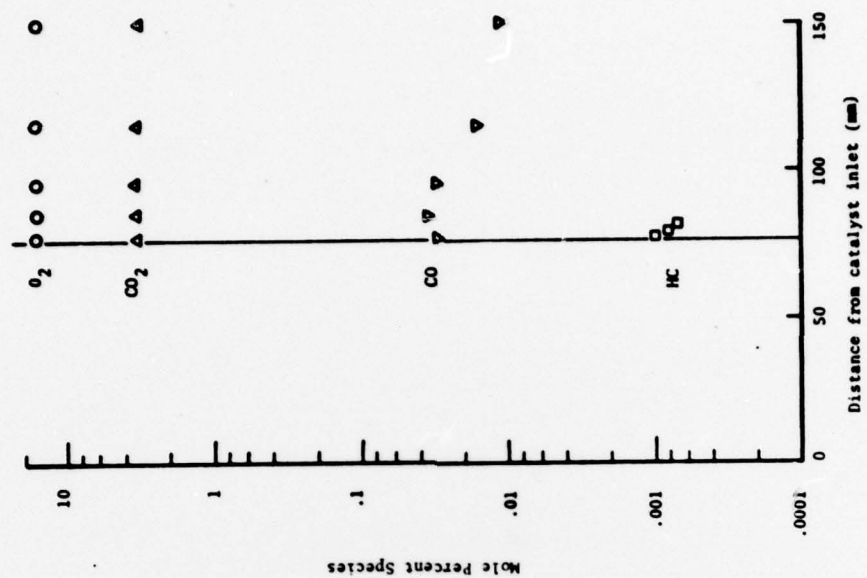


Figure 8

Run 11-2-C₃H₈



Run 11-2-C₃H₈

$P_{in} = 110 \text{ kPa}$

$T_{in} = 750 \text{ K}$

$X = 0.30$

$H_2O = 1.3 \text{ mol \%}$

$u_{ref} = 6 \text{ m/s}$

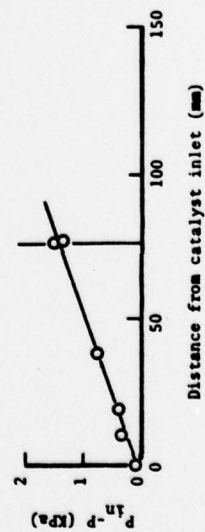
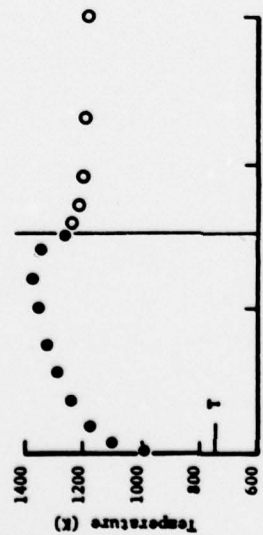


Figure 9

Run 11-13-C₃H₈

Run 11-13-C₃H₈
 $P_{in} = 110 \text{ kPa}$
 $T_{in} = 750 \text{ K}$
 $X = 0.30$
 $H_2O = 1.2 \text{ mol \%}$
 $u_{ref} = 4 \text{ m/s}$

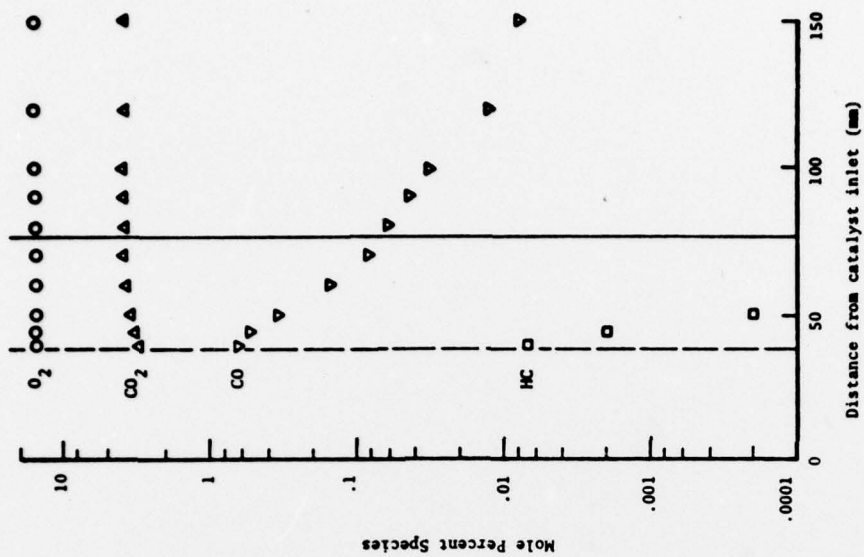
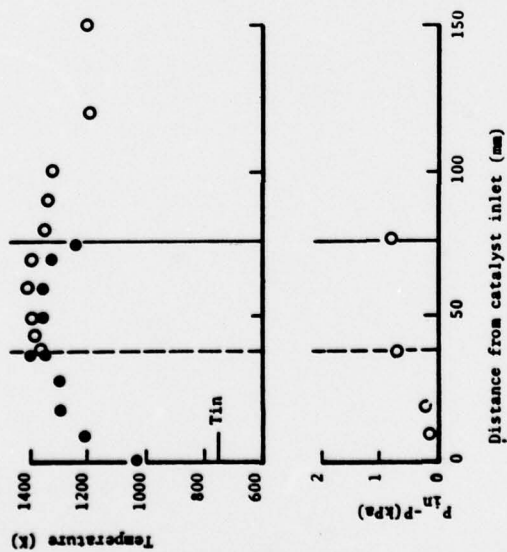


Figure 10